Effective Heat Transfer Parameters for Transient Packed-Bed Models

A general theory is presented for the prediction of effective axial conductivity and overall radial heat transfer coefficient for fluid flow through packed beds, under either transient or steady state conditions. The parameters to be used in each case may be quite different, depending on the dominant phase for heat transfer. The theory unifies all previous results and explains conflicting conclusions arrived at by different workers. The ability of a pseudohomogeneous model to reproduce packed-bed transients is shown to depend upon the choice of effective parameters.

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SCOPE

In many cases the essential behavior of a fixed-bed reactor (location and magnitude of hot-spot, changes in average temperature and composition, etc.) can be represented by one- or two-dimensional pseudohomogeneous models. The use of such models for dynamic reactor studies is desirable when model simplification is important, for example in control or optimal design studies, where much repetitive solution is involved.

The heat transfer parameters in pseudohomogeneous models (for example overall heat transfer coefficient U, effective axial and radial thermal conductivities k_a and k_r, and apparent wall heat transfer coefficient h_w) must represent many physical phenomena, and cannot reliably be obtained from empirical correlations extrapolated from laboratory to plant conditions. For steady state models, it has been shown (Dixon and Cresswell, 1979; Dixon 1985) that it is possible to obtain relations between the effective pseudohomogeneous parameters and the more fundamental parameters of a heterogeneous two-dimensional model that can be reliably correlated and extrapolated. These relations have been used successfully in reactor design and scale-up work (Cropley et al., 1984).

For transient modeling several authors have argued that the same parameters may be used as for steady state work, as in an extensive investigation of the effective axial conductivity in adiabatic fixed beds made by Vortmeyer and coworkers (Vortmeyer and Schaefer, 1974; Vortmeyer and Berninger, 1982; Gunn and Vortmeyer, 1984). Cresswell and Dixon (1982), on the other hand, found that there may be large differences between the effective parameters suitable for dynamic modeling and those suitable for steady state modeling, for the case of an adiabatic bed with negligible solid conduction.

The intention of the present work is to unify and extend all previous results for pseudohomogeneous model effective parameters, for adiabatic and nonadiabetic beds, and for both transient and steady state models. The work provides new relations between single-phase and effective parameters, and shows when these reduce to those of the steady state case, thereby explaining the conclusions of previous writers. The limitations of using pseudohomogeneous models are also mentioned, from the point of view of selection of effective parameters.

CONCLUSIONS AND SIGNIFICANCE

The effective heat transfer parameters used in pseudohomogeneous models of packed-bed heat transfer may be quite different in the steady state and transient cases. In particular, the formulas derived previously for the effective axial thermal conductivity and overall radial heat transfer coefficient for use in steady state

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models (Dixon and Cresswell, 1979; Dixon, 1985) can only be used for transient modeling for low flow rates ($Re < 10$ typically). For high flow rates ($Re > 100$) a rather complicated set of model-matching equations must be solved numerically, to yield values of Pe_a and Nu_o that are considerably reduced from the corresponding steady state values. For the intermediate range graphical interpolation may be used. Dynamic simulations of packed bed thermal response that use the wrong set of parameters will misrepresent the dynamic characteristics of the bed.

The analysis presented here explains why several recent studies of the transient behavior of packed beds were able to use parameter estimates obtained by steady state experimentation—the studies were all carried out at low Re. The simulation of industrial beds, which run at much higher flow rates, would not be so successful. The results of laboratory heat transfer studies have also been misleading, as nearly all data have been taken at low Re and therefore no differences observed between transient and steady state results. The exclusive use of adiabatic beds, for which the differences are much reduced, has also contributed to this misapprehension.

Pseudohomogeneous models cannot both follow the dynamic response and reproduce the final steady state following a step change in fixed-bed inlet temperature, unless the ratio of the fluid phase and solid phase heat capacities is regarded as an effective quantity. Pseudohomogeneous model simulations using an effective heat capacity ratio compare very well with results obtained by numerical solution of the two-phase model equations.

Introduction

Many investigators have found that pseudohomogeneous models of transient heat transfer in packed beds give quite adequate results. Often a one-dimensional single-phase model is all that can be justified due to uncertainty in stoichiometry and/or kinetic parameters (Hoiberg et al., 1971). Several workers have used pseudohomogeneous models for reactor simulation (Hansen and Jorgensen, 1976; Vortmeyer and Jahnel, 1972; Clement and Jorgensen, 1983; Bonvin et al., 1983); others have used these more tractable models for investigation of multiplicity and parameter sensitivity (Hlavacek et al., 1982). For more extreme conditions a two-phase model may be needed, as found by Khanna and Seinfeld (1982) for the highly exothermic methanation reaction, and by Lee and Agnew (1977) even for steady state modeling of vinyl chloride synthesis.

Hoiberg et al. (1971) have stated that an overall heat transfer coefficient U chosen for good dynamic matching of one- and two-phase models would be incompatible with that for steady state. They also found that for transients U should depend on time and axial position to match one- and two-phase models perfectly. This may not be necessary for approximate matching. It was assumed in most other studies that parameters determined from steady state heat transfer experiments could be used directly in dynamic reactor simulations. For example, Clement and Jorgensen (1983) fitted their pseudohomogeneous model to dynamic and steady state data simultaneously, to improve the parameter estimates.

For nonadiabatic steady state simulation, the effective parameters have been given in terms of individual-phase parameters by Dixon and Cresswell (1979) and later in a more complete form by Dixon (1985). In the latter work the relations were shown to have a simple interpretation in terms of heat transfer resistance models. The recommended equations were

$$\frac{1}{Pe_a} = \frac{1}{Pe_{af}} + \frac{k_{as}/k_f}{RePr} \tag{1}$$

$$\frac{1}{Pe_r}\frac{Bi}{Bi+4} = \frac{1}{Pe_{rf}}\frac{Bi_f}{Bi_f+4} + \frac{k_{rs}/k_f}{RePr}\frac{Bi_s}{Bi_s+4} \tag{2}$$

and the limiting conditions

$$Bi \rightarrow \begin{matrix} Bi_f & Re \rightarrow \infty \\ Bi_s & Re \rightarrow 0 \end{matrix} \tag{3}$$

where taking $Bi = Bi_f$ in Eq. 2 gave good predictions of Pe_r over a wide range of Re. For Bi an interpolating form between the two limits was used (Dixon, 1985).

For nonadiabatic dynamic simulation, some partial results were given by Cresswell and Dixon (1982), which will be shown here to correspond to the case of negligible solid conduction, i.e., high flow rate. The present work generalizes the parameter relations for the transient case, and gives our previous results and those of nearly all other workers as special cases.

The exception is the work of Levec and Carbonell (1985a,b) based on the spatial-averaging method. This method leads to a different two-phase model from that considered here, in which extra terms appear representing coupling between convective and dispersive mechanisms. Although it is a more rigorously correct derivation of the transport equations for a porous medium than the shell-balance method, spatial averaging introduces new transport parameters for which there are no experimental or a priori estimates. It has not yet been convincingly demonstrated that the coupling terms containing these parameters represent significant phenomena, although Levec and Carbonell (1985b) argue that they do. Furthermore, the method appears to be restricted to laminar flow in the interstitial channels of the bed, and to an isotropic porous medium. Its application to reactor heat-exchanger tubes, with significant wall effects, operating at industrial flow rates, must therefore await further development.

Model Equations

Consider a nonadiabatic tubular fixed bed with the wall held at a constant temperature T_w, and both phases initially also at a uniform temperature T_w. At time $t = 0$ the entering fluid at $z = 0$ undergoes a step change in temperature to T_0, without change in flow rate. The heat capacity of the wall is taken as negligible (Hoiberg et al., 1971; Lee and Agnew, 1977), and it is desired to

follow the transient response of the bed until a nonuniform steady state is achieved.

The transient one-phase energy balance for the pseudohomogeneous model gives

$$[(1-\epsilon)\rho_s c_s + \epsilon\rho_f c_f]\frac{\partial T_b}{\partial t} = -\dot{m}_f c_f \frac{\partial T_b}{\partial z} + k_a \frac{\partial^2 T_b}{\partial z^2} + k_r\left(\frac{\partial^2 T_b}{\partial r^2} + \frac{1}{r}\frac{\partial T_b}{\partial r}\right) \quad (4)$$

with

$$\frac{\partial T_b}{\partial r} = 0 \quad \text{at } r = 0 \quad (5)$$

$$-k_r \frac{\partial T_b}{\partial r} = h_w(T_b - T_w) \quad \text{at } r = R \quad (6)$$

$$T_b \to T_w \quad \text{as } z \to \infty \quad (7)$$

$$T_b = T_0 \quad \text{at } z = 0 \quad (8)$$

$$T_b = T_w \quad \text{for } t < 0 \quad (9)$$

The equations may be rendered dimensionless in the usual way, and introducing suitable dimensionless groups gives

$$(1+H)\frac{\partial \nu}{\partial \tau} = -\frac{\partial \nu}{\partial x} + \frac{1}{Pe_a}\frac{\partial^2 \nu}{\partial x^2} + \frac{1}{Pe_r}\alpha^2\left(\frac{\partial^2 \nu}{\partial y^2} + \frac{1}{y}\frac{\partial \nu}{\partial y}\right) \quad (10)$$

where time has been scaled by the transformation

$$\tau = t\dot{m}_f c_f/\rho_s c_s d_p(1-\epsilon) \quad (11)$$

for convenience.

The boundary conditions are then

$$\frac{\partial \nu}{\partial y} = 0 \quad \text{at } y = 0 \quad (12)$$

$$\frac{\partial \nu}{\partial y} + Bi\nu = 0 \quad \text{at } y = 1 \quad (13)$$

$$\nu \to 0 \quad \text{as } x \to \infty \quad (14)$$

$$\nu = 1 \quad \text{at } x = 0 \quad (15)$$

$$\nu = 0 \quad \text{for } \tau < 0 \quad (16)$$

The radial derivatives may be discretized by a one-point collocation method (Villadsen and Michelsen, 1978; Dixon and Cresswell, 1979) to yield a one-dimensional model in terms of the temperature at the collocation point ($y_1 = 1/\sqrt{2}$)

$$(1+H)\frac{\partial \nu_1}{\partial \tau} = -\frac{\partial \nu_1}{\partial x} + \frac{1}{Pe_a}\frac{\partial^2 \nu_1}{\partial x^2} - \frac{1}{Pe_r}\frac{8Bi}{Bi+4}\alpha^2\nu_1 \quad (17)$$

For this choice of collocation point ν_1 may be identified as the radial bed average temperature. An overall heat transfer coefficient U based on the difference between wall and average temperatures is defined by

$$\frac{1}{U} = \frac{1}{h_w} + \frac{R/4}{k_r} \quad (18)$$

in which case Eq. 17 is equivalent to starting with a one-dimensional model with a dimensionless overall heat transfer group given by

$$M = \alpha\frac{2U}{\dot{m}_f c_f} = \frac{1}{Pe_r}\left(\frac{8Bi}{Bi+4}\right)\alpha^2 \quad (19)$$

A transient two-phase model, in which both phases are regarded as continuous, may be derived similarly and presented in dimensionless form

$$H\frac{\partial T}{\partial \tau} = -\frac{\partial T}{\partial x} + \frac{1}{Pe_{af}}\frac{\partial^2 T}{\partial x^2} + \frac{1}{Pe_{rf}}\alpha^2\left(\frac{\partial^2 T}{\partial y^2} + \frac{1}{y}\frac{\partial T}{\partial y}\right) + N(\theta - T) \quad (20)$$

$$\frac{\partial \theta}{\partial \tau} = \frac{k_{as}/k_f}{RePr}\frac{\partial^2 \theta}{\partial x^2} + \frac{k_{rs}/k_f}{RePr}\alpha^2\left(\frac{\partial^2 \theta}{\partial y^2} + \frac{1}{y}\frac{\partial \theta}{\partial y}\right) + N(T - \theta) \quad (21)$$

with

$$\frac{\partial T}{\partial y} = \frac{\partial \theta}{\partial y} = 0 \quad \text{at } y = 0 \quad (22)$$

$$\frac{\partial T}{\partial y} + Bi_f T = 0 \quad \text{at } y = 1 \quad (23)$$

$$\frac{\partial \theta}{\partial y} + Bi_s\theta = 0 \quad (24)$$

$$T, \theta \to 0 \quad \text{as } x \to \infty \quad (25)$$

$$T = \theta = 1 \quad \text{at } x = 0 \quad (26)$$

$$T = \theta = 0 \quad \text{for } \tau < 0 \quad (27)$$

One-point collocation and a suitable definition of individual phase overall heat transfer coefficients again leads to a one-dimensional model

$$H\frac{\partial T_1}{\partial \tau} = -\frac{\partial T_1}{\partial x} + \frac{1}{Pe_{af}}\frac{\partial^2 T_1}{\partial x^2} - M_f T_1 + N(\theta_1 - T_1) \quad (28)$$

$$\frac{\partial \theta_1}{\partial \tau} = \frac{k_{as}/k_f}{RePr}\frac{\partial^2 \theta_1}{\partial x^2} - M_s\theta_1 + N(T_1 - \theta_1) \quad (29)$$

The starting point for the following analysis may be taken as either the two-dimensional models (Eqs. 10–16 and 20–27), which are then reduced by one-point collocation, or directly as the one-dimensional models (Eqs. 14–17 and 25–29). In either case, it is assumed that the individual phase transport parameters of the two-phase model are fundamental, and do not change between transient and steady state models. The effective param-

eters of the one-phase model, however, may be built up from the fundamental parameters in different ways, depending upon which features of the two-phase model the one-phase model is required to represent.

Mathematical Development

Further analysis of the model equations proceeds by eliminating the time derivatives by the Laplace transform method, and solving the resulting ODE's for the moments of the transfer function. For the two-phase model exact solution is not easy, and an approximate solution is obtained by perturbation methods.

One-phase model

Taking the Laplace transform of Eq. 17 and setting $G_1(x, s) = s\bar{\nu}_1(x, s)$ gives

$$0 = -\frac{\partial G_1}{\partial x} + \frac{1}{Pe_a}\frac{\partial^2 G_1}{\partial x^2} - (M + s + sH)G_1 \quad (30)$$

which may be solved together with the corresponding boundary conditions to Eqs. 14 and 15 to give

$$G_1(x, s) = \exp\left\{\frac{Pe_a x}{2}[1 - \beta(s)]\right\} \quad (31)$$

where

$$\beta(s) = [1 + 4(M + s + sH)/Pe_a]^{1/2} \quad (32)$$

The zero moment (m_0), first moment (m_1), and second central moment (μ_2) of $G_1(x, s)$ are defined by

$$m_0(x) = \lim_{s\to 0} G_1(x, s) \quad (33)$$

$$m_1(x) = -\frac{1}{m_0}\lim_{s\to 0}\frac{\partial G_1}{\partial s} \quad (34)$$

$$\mu_2(x) = \frac{1}{m_0}\lim_{s\to 0}\frac{\partial^2 G_1}{\partial s^2} - m_1^2 \quad (35)$$

These moments are easily evaluated and are given in Table 1, where $\beta_0 = \lim_{s\to 0}\beta(s) = (1 + 4M/Pe_a)^{1/2}$.

Two-phase model

Taking Laplace transforms of Eqs. 28 and 29 and defining $G_T(x, s) = s\bar{T}_1(x, s)$, $G_\theta(x, s) = s\bar{\theta}_1(x, s)$ gives

$$0 = -\frac{\partial G_T}{\partial x} + \frac{1}{Pe_{af}}\frac{\partial^2 G_T}{\partial x^2} - (M_f + sH)G_T + N(G_\theta - G_T) \quad (36)$$

$$0 = \frac{k_{as}/k_f}{RePr}\frac{\partial^2 G_\theta}{\partial x^2} - (M_s + s)G_\theta + N(G_T - G_\theta) \quad (37)$$

To make further progress the equations must be decoupled, and this is most conveniently done by perturbation methods. It should be noted that the original approach of Dixon and Cresswell (1979) involved perturbation away from zero axial solid conduction, and led to preferential treatment of the fluid phase. In this work consideration of small and large Re leads naturally to perturbation solutions for these equations, in which both phases are treated equally.

Large Re

As $Re \to \infty$, the coefficients M_s and $[k_{as}/k_f/(RePr)]$ tend to zero linearly with Re. For large enough Re, $N \propto Re^{-0.4}$ and Bi_f (and thus M_f) $\propto Re^{-0.4}$ also. Therefore the two solid-phase terms become small first and Eq. 37 may be written

$$0 = \eta\frac{k_{as}/k_f}{RePr}\frac{\partial^2 G_\theta}{\partial x^2} - (\eta M_s + s)G_\theta + N(G_T - G_\theta) \quad (38)$$

Now expanding G_θ in powers of the perturbation parameter η gives

$$G_\theta(\eta, x) = \psi_0(x) + \eta\psi_1(x) + \cdots \quad (39)$$

Substitution of Eq. 39 into Eq. 38 and equating coefficients of powers of η gives

$$\psi_0(x) = \frac{N}{N + s}G_T(x) \quad (40)$$

and

$$\psi_1(x) = \frac{k_{as}/k_f}{RePr}\frac{N}{(N + s)^2}\frac{\partial^2 G_T}{\partial x^2} - \frac{M_s N}{(N + s)^2}G_T(x) \quad (41)$$

Table 1. First Three Moments of the One- and Two-Phase Models

Moment	One-phase Model	Two-phase Model (Fluid phase, $Re \to \infty$)	Two-phase Model (Solid phase, $Re \to 0$)
m_0	$\exp\left[\frac{Pe_a x}{2}(1 - \beta_0)\right]$	$\exp\left[\frac{P_0 x}{2}(1 - B_0)\right]$	$\exp\left[\frac{P_0 x}{2}(1 - B_0)\right]$
m_1	$\frac{x}{\beta_0}(1 + H)$	$\frac{x}{2}[P_0 B_{10} - P_{10}(1 - B_0)]$	$\frac{x}{B_0}(1 + H)$
μ_2	$\frac{2(1 + H)^2 x}{Pe_a \beta_0^3}$	$\frac{x}{2}[(1 - B_0)P_{20} - 2P_{10}B_{10} - P_0 B_{20}]$	$\frac{2(1 + H)^2 x}{P_0 B_0^3}$

These are substituted into the expansion in Eq. 39, which is truncated after the first two terms. Then $\eta = 1$ recovers the original problem, and gives

$$G_\theta(x) \simeq \frac{k_{as}/k_f}{RePr}\frac{N}{(N+s)^2}\frac{\partial^2 G_T}{\partial x^2} + \frac{N}{N+s}\left(1 - \frac{M_s}{(N+s)}\right)G_T(x) \quad (42)$$

Now substitution into Eq. 36 for $G_\theta(x)$ gives an equation of the form

$$0 = -\frac{\partial G_T}{\partial x} + \frac{1}{P(s)}\frac{\partial^2 G_T}{\partial x^2} - F(s)G_T \quad (43)$$

with solution

$$G_T(x, s) = \exp\left\{\frac{P(s)x}{2}[1 - B(s)]\right\} \quad (44)$$

where

$$\frac{1}{P(s)} = \frac{1}{Pe_{af}} + \frac{k_{as}/k_f}{RePr}\frac{N^2}{(N+s)^2} \quad (45)$$

$$F(s) = M_f + sH + \frac{sN}{N+s} + \frac{N^2}{(N+s)^2}M_s \quad (46)$$

and

$$B(s) = [1 + 4F(s)/P(s)]^{1/2} \quad (47)$$

The first three moments of $G_T(x, s)$ may now be obtained after some tedious algebra; the results are again given in Table 1.

Small Re

Multiply Eq. 36 through by $(RePr)$ to obtain

$$0 = -(RePr)\frac{\partial G_T}{\partial x} + \frac{k_{af}}{k_f}\frac{\partial^2 G_T}{\partial x^2} - \left[\alpha^2\frac{k_{rf}}{k_f}\frac{8Bi_f}{Bi_f + 4} + sH(RePr)\right]G_T + N(RePr)(G_\theta - G_T) \quad (48)$$

and consider limits as $Re \to 0$. In that limit k_{af}/k_f and k_{rf}/k_f both tend to ϵ, and $8Bi_f/(Bi_f + 4) \to 8$. It can be shown that $N(RePr) \simeq 6(1 - \epsilon)Nu_{fs}$, and as $Re \to 0$ recent work on interphase heat transfer (Wakao et al., 1979) suggests that $Nu_{fs} \simeq 2$. Then the interphase transfer term in Eq. 48 dominates and the equation may be written (setting $\eta = RePr$)

$$0 = \eta\left[-\frac{\partial G_T}{\partial x} + \frac{1}{Pe_{af}}\frac{\partial^2 G_T}{\partial x^2} - (M_f + sH)G_T\right] + 6(1 - \epsilon)Nu_{fs}(G_\theta - G_T) \quad (49)$$

Expanding $G_T(\eta, x) = \phi_0(x) + \eta\phi_1(x) + \cdots$ (50)

substituting into Eq. 49 and equating coefficients of powers of η leads to

$$\phi_0(x) = G_\theta(x) \quad (51)$$

and

$$\phi_1(x) = \frac{1}{6(1 - \epsilon)Nu_{fs}} \cdot \left[-\frac{\partial G_\theta}{\partial x} + \frac{1}{Pe_{af}}\frac{\partial^2 G_\theta}{\partial x^2} - (M_f + sH)G_\theta\right] \quad (52)$$

These are substituted into Eq. 50, and terms through the first order in η only are retained. Then $\eta = (RePr)$ recovers the original problem and gives

$$G_T(x) \simeq G_\theta(x) + \frac{1}{N}\left[-\frac{\partial G_\theta}{\partial x} + \frac{1}{Pe_{af}}\frac{\partial^2 G_\theta}{\partial x^2} - (M_f + sH)G_\theta\right] \quad (53)$$

Now substitution into Eq. 37 for $G_T(x)$ gives

$$0 = \left(\frac{k_{as}/k_f}{RePr} + \frac{1}{Pe_{af}}\right)\frac{\partial^2 G_\theta}{\partial x^2} - \frac{\partial G_\theta}{\partial x} - (M_f + M_s + sH + s)G_\theta \quad (54)$$

Clearly this equation becomes identical to that for the one-phase model, Eq. 30, if

$$\frac{1}{Pe_a} = \frac{1}{Pe_{af}} + \frac{k_{as}/k_f}{RePr} \quad (1)$$

and

$$M = M_f + M_s \quad (55)$$

which are just the steady state model-matching relations, when the dimensionless overall heat transfer groups are expanded. Thus all moments of the one-phase and two-phase models agree in the limit $Re \to 0$, with the above choices of effective parameters. For completeness, the first three moments are given in Table 1 for this case also.

Relationships Among Model Parameters

Inspection of the entries in Table 1 shows that the zeroth moments of the one-phase and two-phase models may be matched for all cases by setting

$$Pe_a(1 - \beta_0) = P_0(1 - B_0) \quad (56)$$

and choosing $Pe_a = P_0$, $\beta_0 = B_0$ yields precisely the steady state relations, Eqs. 1 and 2, as expected. These relations may also be obtained by direct analysis of the steady state equations, and are equivalent to assuming equal radial average temperatures in the two phases, and requiring

$$v_1(x) = T_1(x) = \theta_1(x) \quad (57)$$

for the homogeneous bed radial average temperature. In order to obtain the steady state relation of Eq. 3, a further condition was imposed on the radial profiles

$$v(x)|_{y=1} = \begin{cases} T(x)|_{y=1} & (Re \to \infty) \\ \theta(x)|_{y=1} & (Re \to 0) \end{cases} \tag{58}$$

noting that one-point collocation leads to representation of the radial profile by a quadratic interpolant with nodes at $y = y_1$ and $y = 1$. Thus the entire approximate radial profile was matched by Eqs. 57 and 58. The equivalent in the present analysis would be to develop a transfer function $G'(s)$ for the response at $y = 1$, and to match the zero moment $m_0'(x)$ of the homogeneous bed response to the zero moment of an appropriate individual-phase response, all at $y = 1$. This again leads to the separate relation for Bi in Eq. 3.

To match the higher-order moments m_1 and μ_2, the two cases of high and low Re must be considered separately. For low Re Table 1 again shows that both higher-order moments may be matched by the steady state relations embodied in Eq. 56, so that in fact all three moments may be simultaneously matched in this case. For high Re, matching m_1 and μ_2 leads to

$$\frac{1+H}{\beta_0} = \frac{1}{2}[P_0 B_{10} - P_{10}(1 - B_0)] \tag{59}$$

and

$$\frac{2(1+H)^2}{Pe_a \beta_0^3} = \frac{1}{2}[(1 - B_0)P_{20} - 2P_{10}B_{10} - P_0 B_{20}] \tag{60}$$

These relations are extremely complicated, and cannot be simplified further. They must be solved numerically for Pe_a and β_0 (and thence M), noting that all quantities on the righthand sides may be obtained from individual-phase parameters, as shown in Table 2. Some computational results are given in the following section.

In matching the higher-order moments m_1 and μ_2, it seems natural to try to obtain relations for Pe_r and Bi separately, analogously to the steady state case discussed above. It may soon be shown, however, that m_1 and μ_2 are independent of radial position, and so provide no further matching criterion. Only for the low-Re case, where the zero-order moments are also matched, can Bi and Pe_r be separated, and the result is as for the steady state case.

Although the general relations of Eqs. 59 and 60 are intractable, some special cases are of interest.

Negligible Axial and Radial Solid Conduction. This case was originally considered by Cresswell and Dixon (1982). Let M_s, $k_{as}/k_f \to 0$, then the general high-Re relations simplify to

$$\frac{1}{Pe_a} = \frac{1}{N(1+H)^2} + \frac{1}{Pe_{af}}\left(1 + \frac{4M_f}{N(1+H)^2}\right) \tag{61}$$

and

$$M = \frac{M_f}{\left[1 + \dfrac{4M_f + Pe_{af}}{N(1+H)^2}\right]} \tag{62}$$

Note that Eq. 61 is a corrected version of the earlier result, where the second division by N was inadvertently omitted.

Adiabatic Bed with Gas Flow. This case has received most attention in the literature. Take $M_f = M_s = 0$ and set $H \simeq 0$, then

$$\frac{1}{Pe_a} = \frac{1}{Pe_{af}} + \frac{k_{as}/k_f}{RePr} + \frac{1}{N} \tag{63}$$

which is the formula discussed previously by Vortmeyer and Berninger (1982).

Adiabatic Bed with any Fluid. In this case $H \neq 0$, and the formula is

$$\frac{1}{Pe_a} = \frac{1}{Pe_{af}} + \frac{k_{as}/k_f}{RePr} + \frac{1}{(1+H)^2 N} \tag{64}$$

which corresponds to the result obtained by Vortmeyer and

Table 2. Expressions Appearing in the Moments of Table 1

Quantity	Notation	Full Expression
$P\|_{s=0}$	P_0	$\left[\frac{1}{Pe_{af}} + \frac{k_{as}/k_f}{RePr}\right]^{-1}$
$B\|_{s=0}$	B_0	$\left[1 + \frac{4}{P_0}(M_f + M_s)\right]^{1/2}$
$\left.\frac{dP}{ds}\right\|_{s=0}$	P_{10}	$2\alpha P_0^2 \frac{k_{as}/k_f}{RePr}\frac{1}{N}$
$\left.\frac{dB}{ds}\right\|_{s=0}$	B_{10}	$\frac{2\alpha}{B_0}\left[\frac{1}{P_0}\left(1 + H - 2\frac{M_s}{N}\right) - 2\frac{k_{as}/k_f}{RePr}(M_f + M_s)\frac{1}{N}\right]$
$\left.\frac{d^2P}{ds^2}\right\|_{s=0}$	P_{20}	$2\alpha^2 P_0^2 \frac{k_{as}/k_f}{RePr}\frac{1}{N^2}\left(4P_0\frac{k_{as}/k_f}{RePr} - 3\right)$
$\left.\frac{d^2B}{ds^2}\right\|_{s=0}$	B_{20}	$\frac{2\alpha^2}{B_0 N}\left[\frac{1}{P_0}\left(6\frac{M_s}{N} - 2\right) + 6\frac{k_{as}/k_f}{RePr}(M_f + M_s)\frac{1}{N} - 4\frac{k_{as}/k_f}{RePr}\left(1 + H - 2\frac{M_s}{N}\right)\right] - \frac{B_{10}^2}{B_0}$

Schaefer (1974). This equation may also be obtained from the spatial-averaging analysis of Levec and Carbonell (1985a,b), if the dispersion-convection coupling terms are neglected.

Negligible Axial Fluid Dispersion and Axial Solid Conduction. Let k_{af}, $k_{as} \rightarrow 0$ in the two-phase model. For low Re, or zero-moment matching, the effective parameters are given by

$$Pe_a = \infty \tag{65}$$

and

$$M = M_f + M_s \tag{66}$$

so that the one-phase model has no effective axial dispersion ($k_a = 0$).

For high Re the first and second fluid-phase moments are matched by the relations

$$\frac{1 + H}{\beta_0} = \left(1 + H - \frac{2M_s}{N}\right) \tag{67}$$

and

$$\frac{(1 + H)^2}{Pe_a \beta_0^3} = \frac{1}{N}\left(1 - \frac{3M_s}{N}\right) \tag{68}$$

which can be solved for Pe_a and M. The interesting point is that a finite value is obtained for Pe_a, corresponding to nonzero effective axial thermal conductivity. This is due to the contribution by interphase transfer, which acts as an effective axial dispersion mechanism. Thus, an axially dispersed one-phase model is needed to reproduce the transient characteristics of a two-phase model, even though the axial dispersion and conduction terms are not explicitly included in the two-phase model.

Discussion of Parameter Relationships

There are considerable differences between the parameter relations applicable to steady state modeling (matching of m_0) and those applicable to dynamic or transient modeling (matching of m_1 and μ_2). In the former case the pseudohomogeneous model is required to reproduce the final steady state profile of the bed, and to the order of approximation used here the fluid and solid phases have the same radial-average temperature. Thus a single relation is obtained in which the interphase transport parameter does not appear. In the latter case, the pseudohomogeneous model is required to reproduce characteristics of the transient response, and the final steady states may be poorly matched. For low flow rates where transport through the fluid phase is mainly by slow molecular mechanisms, the dynamic response of the bed as a whole will be that of the high-heat-capacity solid, and will be slow. The transient will propagate through the bed slowly enough for interphase heat transfer to keep the two phases at similar temperature, leading to the same parameter relations as for the steady state case. For high flow rates with fast turbulent fluid phase transport, the dynamic response of the bed will be that of the fluid, with the solid lagging behind, and a large temperature difference can exist between the phases. In this case interphase transport will have a large influence on the bed dynamics, as is seen in the parameter relations, which no longer reflect equality of the average temperatures.

The parameter relations for the effective axial Peclet number Pe_a, and the overall bed Nusselt number $Nu_0 (= MRePr/2\alpha)$ are shown in Figures 1 and 2. Typical conditions were chosen as a gas flowing through a low conductivity packing, leading to $H \simeq 0$, $k_{rs}/k_f = k_{as}/k_f = 8$ and $d_t/d_p = 10$. The individual phase parameters were obtained from published correlations (Dixon et al., 1984; Melanson and Dixon, 1985; Dixon and LaBua, 1985).

Both Pe_a and Nu_0 are strongly decreased from the steady state formula predictions for $Re > 100$. The high-Re transient relations should be used in this range. The steady state formulas appear to be valid up to $Re = 5$, then there is a transition region in which empirical curve-fitting or graphical methods are needed. The effects of solid conductivity and d_t/d_p were fairly small. For a liquid fluid ($H \simeq 1$) the differences between steady state and transient formulas were somewhat decreased, but still significant.

The overall Nusselt number increases by only 10% with a tenfold increase in Re from 100 to 1,000, however Pe_a undergoes an order-of-magnitude decrease over the same range. Thus k_a

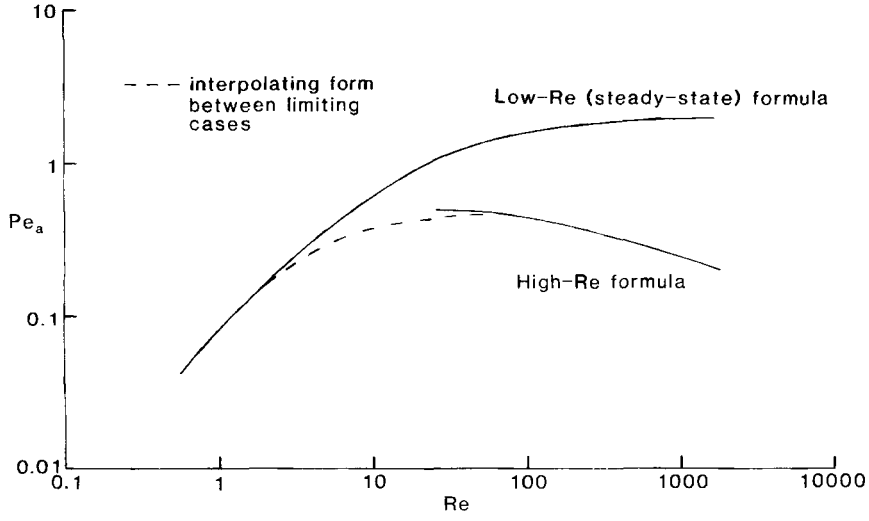

Figure 1. Comparison of steady state and dynamic relations for effective axial Peclet numbers.

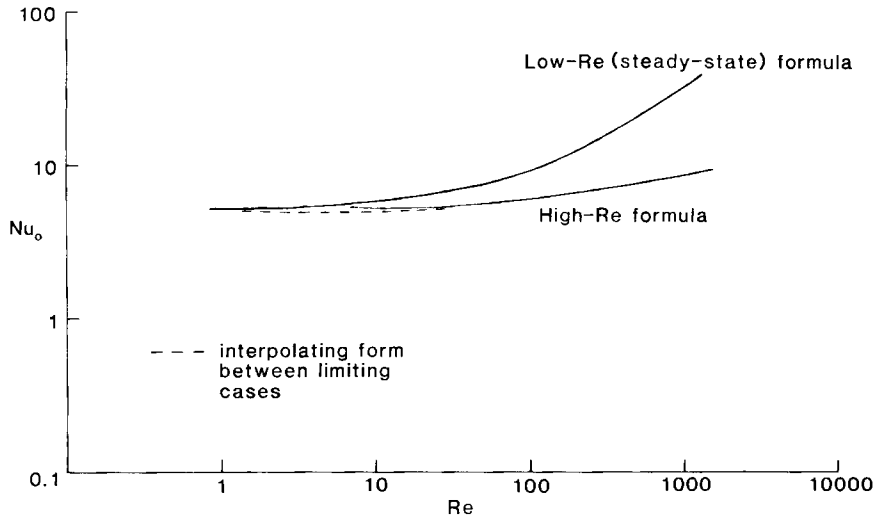

Figure 2. Comparison of steady state and dynamic relations for overall heat transfer Nusselt number.

increases by an order of magnitude over that range. Nu_0 must be used together with Pe_a, and not separately, otherwise predictions of the temperature profile will be extremely sensitive to flow rate. The authors are grateful to one of the reviewers for raising this point.

Comparison of the effective parameter predictions with data is difficult, as the only parameter determinations for the transient case have been for Pe_a under adiabatic conditions. Data from several studies are shown in Figure 3, together with steady state and adiabatic transient prediction formulas. Note that for the adiabatic case the high and low Re asymptotic curves blend more smoothly than for the nonadiabatic case.

In a similar comparison Gunn and Vortmeyer (1984) found that empirical correlations for steady state data by Vortmeyer and Adam (1984) lay close to the transient data, as did the steady state prediction formula (Eq. 1) with theoretical estimates for the fluid-phase contribution. Most of the data, however, lie in the range $Re < 100$, where there should be little or no difference between steady state and transient cases. The only data taken at high enough Re to discriminate between the formulas are those of Goss and Turner (1971) and Turner and Otten (1973). These data show a strong downturn for $Re >$ 1,000 and conclusively show the need for the transient-matching equations at high Re, and the inclusion of an interphase transport term.

Other workers have also assumed that effective parameters are the same for steady and transient models. Clement and Jorgensen (1983) ran their experiments at $Re = 10$, and thus were able to estimate effective parameters simultaneously from steady state and dynamic experiments, as they are the same at such low Re. Similarly Bonvin et al. (1983) used the pseudohomogeneous model for control studies of the autothermal water-gas shift reaction in a fixed bed, and used steady state correlations for Pe_r and Bi to get U. Their reported conditions give $Pe =$ 18, so again this would be permissible. Finally, in a stagnant bed ($Re = 0$) Kaguei et al. (1977) have shown that the effective axial conductivity is the same under steady or transient conditions, although their analysis was for a discrete particle model.

The parameter relations developed here explain the trends of the limited data available, and also explain why previous work-

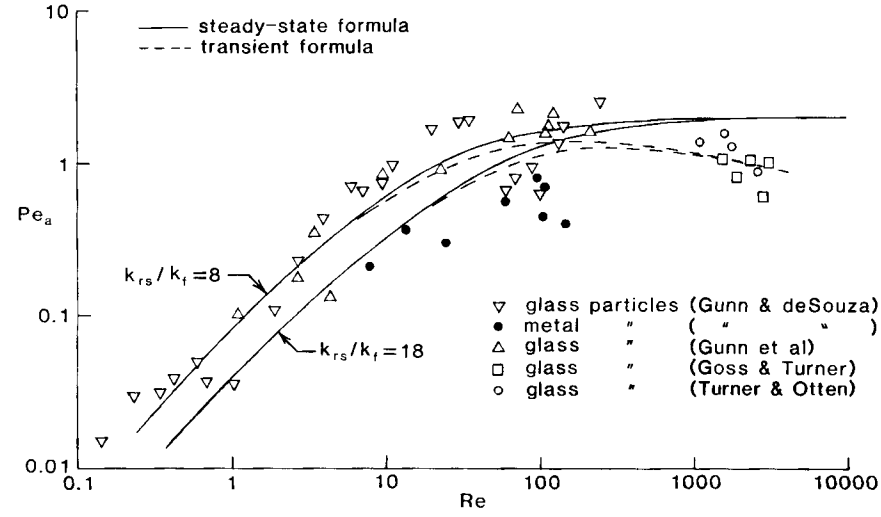

Figure 3. Comparison of prediction formulas with data for the adiabatic effective axial Peclet number.

ers did not observe discrepancies in their results. Transient simulations for the very much higher Re of industrial practice may be greatly in error, however, if the wrong choice of effective parameters is made.

Transient Model Simulations

The one-dimensional pseudohomogeneous model (Eqs. 14–17) may be solved exactly for the radial bed average temperature $\nu_1(x, \tau)$ by the Laplace transform method (i.e., by taking the inverse transform of Eqs. 31 and 32). The response to a step change in inlet temperature is given by

$$\nu_1(x,\tau) = \frac{1}{2}\exp\left(\frac{Pe_a x}{2}\right)\left[\exp(-x\sqrt{a_1 a_2})\,\mathrm{erfc}\left(\frac{x}{2\sqrt{\tau/a_2}} - \sqrt{a_1\tau}\right) + \exp(x\sqrt{a_1 a_2})\,\mathrm{erfc}\left(\frac{x}{2\sqrt{\tau/a_2}} + \sqrt{a_1\tau}\right)\right] \quad (69)$$

where $a_1 = [Pe_a/4 + 2\alpha Nu_0/(RePr)]$, $a_2 = Pe_a(1 + H)$. The one-dimensional two-phase model (Eqs. 25–29) was solved numerically by a method-of-lines finite-difference method, as implemented in the DSS/2 software package (Schiesser, 1976). The fluid and solid phase transient responses were then compared to the pseudohomogeneous model predictions, using effective parameters obtained from the steady state relations (Eqs. 1–2), and from the transient relations (Eqs. 59–60). The results of the comparison are shown in Figure 4.

The responses shown in Figure 4 show the same trends as qualitatively discussed by Cresswell and Dixon (1982). At the moderate value of Re chosen, the solid-phase response lags behind that of the fluid phase, however the two phases approach equality of temperature as steady state is approached. The transient-matching pseudohomogeneous response shows considerable offset from the final steady state, whereas the steady state-matching response shows a much steeper breakthrough curve, with the response being delayed, but reproduces the final steady state well. Thus no choice of the two effective parameters Pe_a and Nu_0 can cause the pseudohomogeneous model to reproduce both steady state and dynamic characteristics of the bed response.

With the above observations in mind, notice that for high Re the effective parameters have been adjusted to match either m_0 or m_1 and μ_2. It is the inability of the transient matching relations to also match the zero-order moments that leads to the offset from the final steady state. In order to match all three moments, an extra adjustable parameter is needed. This is forthcoming if it is agreed to regard H in the pseudohomogeneous model as an effective parameter, rather than a fixed quantity. This means that the model uses an effective heat capacity ratio or, equivalently, an effective fluid phase heat capacity.

Model matching for high Re now means that Eqs. 56, 59, and 60 are satisfied simultaneously, with H replaced by H_{eff} on the lefthand sides of Eqs. 59 and 60. Solving the relations stepwise gives

$$Pe_a = P_0(1 - B_0) + \frac{[P_0B_{10} - P_{10}(1 - B_0)]^2}{[(1 - B_0)P_{20} - 2P_{10}B_{10} - P_0B_{20}]} \quad (70)$$

$$1 + H_{eff} = \frac{4}{Pe_a}\frac{[P_0B_{10} - P_{10}(1 - B_0)]^3}{[(1 - B_0)P_{20} - 2P_{10}B_{10} - P_0B_{20}]} \quad (71)$$

$$\left[1 + \frac{4}{Pe_a}\left(\frac{2\alpha Nu_0}{RePr}\right)\right] = \frac{2(1 + H_{eff})}{[P_0B_{10} - P_{10}(1 - B_0)]} \quad (72)$$

which may be used to evaluate Pe_a, H_{eff}, and Nu_0 sequentially. For low Re the steady state relations apply as usual, with $H_{eff} = H$.

The predicted response from the pseudohomogeneous model with effective parameters given by the hybrid relations, Eqs. 70–72, is shown in Figure 5, again compared to numerical solution of the two-phase model equations. It may be seen that now the one-phase model response agrees well with the final steady state, i.e., there is little or no offset, and also agrees well with the initial breakthrough of the solid phase response, both in location and shape of curve. The approach to steady state is also in good agreement as to shape with fluid and solid phase curves.

The correlations for the effective parameters also undergo some changes due to the different matching formulas. Tha axial Peclet number Pe_a is almost unchanged from the values appropriate for high Re transient matching, shown earlier in Figure 1. The overall Nusselt number correlation, however, now gives values that are almost identical to those from steady state matching. This is reasonable when it is realized that Nu_0 has a large influence on the final steady state, whereas Pe_a has more

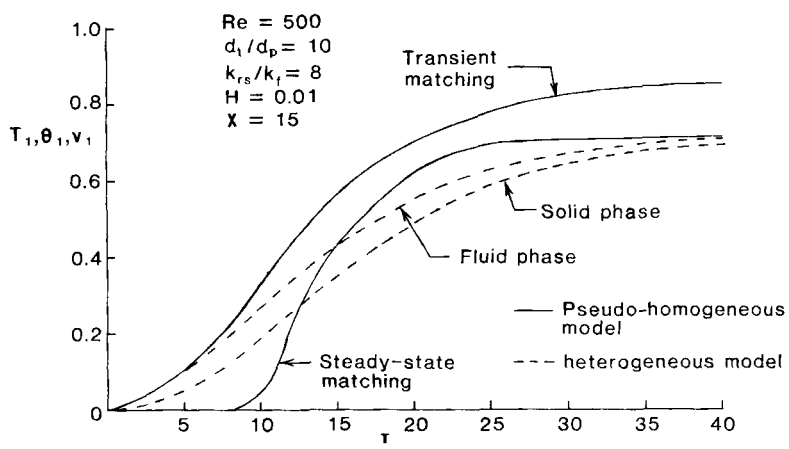

Figure 4. Response curves for two-phase model and different choices of effective parameters in the one-phase model.

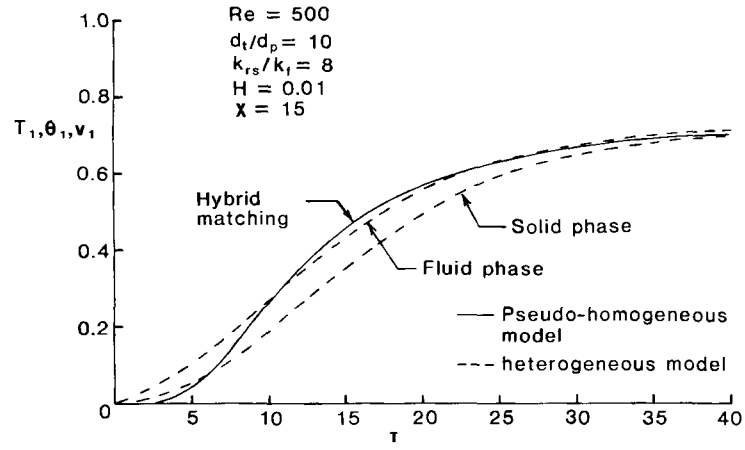

Figure 5. Response curves for two-phase model and hybrid effective parameters in the one-phase model.

influence on the shape or spread of the transient response. The effective heat capacity ratio H_{eff} is shown in Figure 6; the high Re formulas give rather unreliable values for $Re < 50$, and any smooth transition between the high Re effective value and the low Re true value of H has to be speculative. An added bonus in matching all three moments is that it is possible then to match the zero-order moments at the boundary, as discussed previously, which leads to Eq. 3 and a rational basis for choosing Pe_r and Bi for the two-dimensional pseudohomogeneous model.

Notation

All heat transfer parameters defined with respect to total area (void and nonvoid) normal to the direction of heat transfer.

a = specific interfacial surface area
c_f = fluid specific heat
c_s = solid specific heat
d_p = particle diameter
d_t = tube diameter
h = apparent interphase heat transfer coefficient
h_w = apparent wall heat transfer coefficient
k_a = effective axial conductivity
k_r = effective radial conductivity
k_f = fluid molecular conductivity
k_{as} = axial conductivity of the solid
k_{rs} = radial conductivity of the solid
$\dot{m}_f$ = superficial fluid mass velocity
r = tube radial coordinate
R = tube radius
t = time
T_b = bed temperature (one-phase model)
T_w = wall temperature
T_0 = bed inlet temperature
U = overall radial heat transfer coefficient
z = tube axial coordinate

Dimensionless parameters

a_1 = defined after Eq. 65
a_2 = defined after Eq. 65
Bi = apparent wall Biot number, $h_w R/k_r$
Bi_f = fluid-phase wall Biot number, $h_{wf} R/k_{rf}$
Bi_s = solid-phase wall Biot number, $h_{ws} R/k_{rs}$
$B(s)$ = defined in Eq. 47
B_0, B_{10}, B_{20} = defined in Table 2
$F(s)$ = defined in Eq. 46
G, G_1 = transfer function for v; radial-average transfer function
G_T, G_θ = fluid, solid phase transfer functions
H = heat capacity ratio, $\epsilon\rho_f c_f/(1-\epsilon)\rho_s c_s$
H_{eff} = effective heat capacity ratio
m_0, m_1 = zero-order and first-order moments, Eqs. 33–34
M = radial heat transfer group, Eq. 19

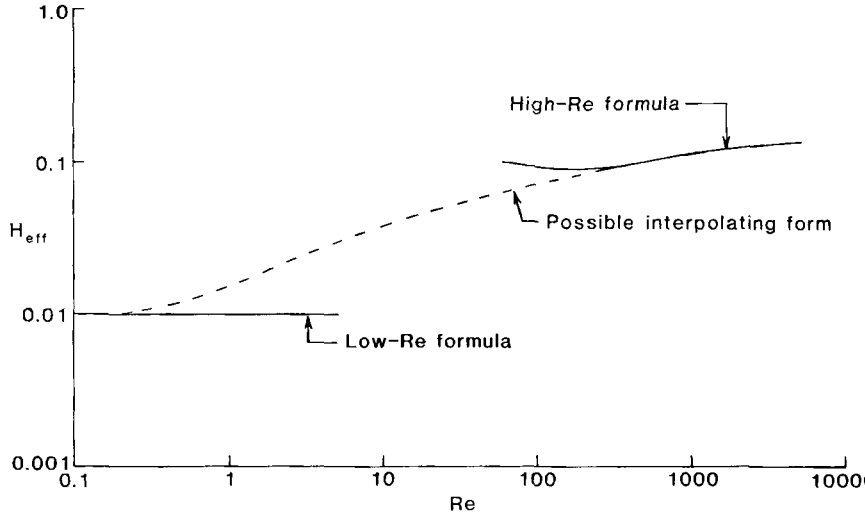

Figure 6. Effective heat capacity ratio.

M_f = fluid-phase radial heat transfer group
M_s = solid-phase radial heat transfer group
N = interphase heat transfer group, $ahd_p/\dot{m}_f c_f$
Nu_{fs} = interphase Nusselt number, hd_p/k_f
Nu_0 = overall Nusselt number, Ud_p/k_f
$P(s)$ = defined in Eq. 45
P_0, P_{10}, P_{20} = defined in Table 2
Pe_a = effective axial Peclet number, $\dot{m}_f c_f d_p/k_a$
Pe_r = effective radial Peclet number, $\dot{m}_f c_f d_p/k_r$
Pe_{af} = fluid-phase axial Peclet number, $\dot{m}_f c_f d_p/k_{af}$
Pe_{rf} = fluid-phase radial Peclet number, $\dot{m}_f c_f d_p/k_{rf}$
Pr = Prandtl number, $\mu c_f/k_f$
Re = Reynolds number, $\dot{m}_f d_p/\mu$
T, T_1 = dimensionless fluid temperature, $(T_f - T_w)/(T_0 - T_w)$; average fluid temperature
x = dimensionless axial coordinate, z/d_p
y = dimensionless radial coordinate, r/R

Greek letters

α = bed aspect ratio, d_p/R
β, β_0 = defined in Eq. 32
ϵ = bed voidage
ϕ = perturbation expansion function
ψ = perturbation expansion function
θ, θ_1 = dimensionless solid temperature, $(T_s - T_w)/(T_0 - T_w)$; average solid temperature
ρ_s = solid phase density
ρ_f = fluid phase density
θ = dimensionless time, $\dot{m}_f c_f t/(1-\epsilon)\rho_s c_s d_p$
ν, ν_1 = dimensionless bed temperature, $(T_b - T_w)/(T_0 - T_w)$; average bed temperature
η = perturbation parameter
μ = fluid-phase viscosity
μ_2 = second-order moment, Eq. 35

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